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Determination of hand soil loading, soil transfer, and particle size variations after hand-pressing and hand-mouthing activities.

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Abstract

Hand-pressing trials and hand-to-mouth soil transfer experiments were conducted to better understand soil loadings, soil transfer ratios for three mouthing activities, and variations in particle size distributions under various conditions. Results indicated that sand caused higher soil loadings on the hand than clay. When the moisture level of clay soil exceeded its liquid limit, soil loadings also increased. Greater pressing pressures also led to larger clay loadings. Clay with a moisture content close to its plastic limit caused the smallest soil loadings due to strong soil cohesion. Particle sizes of the transferred clay were larger than that of the original clay, indicating that hand-pressing and the pressure exerted may have enhanced clay particles of larger sizes adhering onto the hand. Nevertheless, the sizes of most particles that adhered to the hand were still smaller than 150 μm. Higher pressing pressures and greater moisture contents resulted in larger soil loadings on the hand, and transfer ratios became smaller. Transfer ratios from palm-licking with clay particles were smaller than those from finger-mouthing, which may have been due to finer particles that more readily adhered to the skin of the palm and that were transferred from the hand to the mouth with greater difficulty.

Keywords

Exposure; Health risk; Mouthing activity; Soil loading; Soil texture; Soil transfer

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Appendix A. Supplementary data

1. Introduction

A human health risk assessment is the process of estimating the nature and likelihood of adverse health effects in humans resulting from present or anticipated exposures to chemicals in contaminated environmental media (USEPA, 2004). An assessment of exposures to chemicals from contaminated soil via ingestion and dermal contact pathways requires that information be gathered on soil adherence to the skin and hand-to-mouth transfer. These parameters are then used to determine the magnitude of the exposure risk, along with data on the content and speciation of contaminants found in the soil through a chemical analysis.

Children may experience higher exposure risks through soil ingestion and dermal contact than adults, because children exhibit higher frequencies and durations of mouthing behaviors, especially early in their lives. Children also spend more time playing outside on the ground than do adults. Moreover, toxic substances in the soil may pose higher risks to children, because children are more susceptible than adults leading to increased contact with soil and sands. In a US Environmental Protection Agency (EPA) report, children within the age group of 3 to <6 years old were identified as being of great concern, because of relevant types of exposure behaviors that occur in this age range. In particular, children in this age group exhibit high hand-to-mouth activities, which contribute to elevated soil and dust ingestion rates and exposure to contaminants in the soil and dust (USEPA, 2005). Children older than 6 years begin wearing adult-style clothing, which may reduce the available body area for dermal contact, and also their hand-to-mouth activities become less frequent. It was shown that oral contact with the hands and objects, as well as their dermal contact with surfaces decrease for children who are 6 to <11 years old (USEPA, 2005).

Variations in soil ingestion intake due to mouthing behaviors of children depend on the amount of adherence of soil to the skin and the amounts of soil transferred from the object contacted to the skin and mouth. These variables are highly correlated with the size of soil particles and moisture content of the soil (Bergstrom et al., 2011; Choate et al., 2006a; Choate et al., 2006b; Ferguson et al., 2009a; Kissel et al., 1996a; Kissel et al., 1996b; Vasiluk et al., 2011). Sheppard and Evenden (1994) found that soils with particle sizes of <50 μm in diameter preferentially adhered to the skin; the concentrations of soil-bound contaminants, such as lead, mercury, uranium, and hexachlorobenzene, were up to 10-fold greater in the fine adhering fraction of the soil compared to those in bulk soils. Luo et al. (2011) also noted that concentrations of trace metals tended to be greater in the fine fractions of soil than in bulk soils. Moreover, these trace metals were found to mainly accumulate in clay and fine silt.

The contact pressure and duration are also among factors that can influence soil adherence values. Ferguson et al. (2009b) indicated that soil transfer increased as the contact pressure increased, but when the contact durations exceeded 30 s, adherence values did not seem to further increase. Other studies showed that greater test pressures and longer contact durations caused greater soil adherence (Ferguson et al., 2009a, Ferguson et al., 2009b).

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Thus, a better understanding of particle size distributions prior to and after soil transfer from objects to the skin and from skin to the mouth is as important as knowing subsequent amounts of soil transfer for assessing exposures and chemical risks resulting from contacting contaminated soils. Earlier laboratory experiments in which adult subjects pressed their hands onto soil were mainly designed to measure soil adherence values. Results from those studies showed that finer soil particles adhered more efficiently to hands than did coarse soil samples (Ferguson et al., 2009a; Yamamoto et al., 2006). However, available studies pertaining to understanding hand-to-mouth transfer of soils are still limited. The only identifiable study was conducted by Kissel et al. (1998a), in which a laboratory-based examination was performed. The soil tested was a sub-2-mm fraction of natural loamy sand, which was first autoclaved and stored at room temperature under foil. Moisture contents in that experiment ranged 0.8%~1.6%. The experimental results also showed that palm-licking caused the largest amount of hand-to-mouth transfer of soil, followed by finger-mouthing and thumb-sucking (Kissel et al., 1998a).

In this study, hand-pressing trials and hand-to-mouth soil transfer experiments were carried out in an attempt to better understand soil loading on the hands and the tendency of particle size variations during soil-to-hand and hand-to-mouth transfer. We also investigated the dependence of soil transfer, soil loading, and hand-to-mouth transfer ratios during three types of mouthing activities and variations in the particle size distribution due to several parameters, including the soil texture, moisture content, contact time, and pressing pressure.

Recent studies conducted in Taiwan, similar to research reported by Kissel et al. (1998a), were previously undertaken in order to enhance our earlier children behavioral studies pertaining to mouthing activities and soil-ingestion rates of children (Chien et al., 2017; Tsou et al., 2015). In the current study, we aimed to enhance and strengthen the local database established for assessing exposures and health risks of children living near contaminated sites. This area of research has been a key area of concern of the Taiwan Environmental Protection Agency (TEPA, 2010).

2. Experimental

2.1. Test parameters for soil-transfer studies and hand-to-mouth transfers

Hand-pressing trials and hand-to-mouth soil transfer experiments were conducted with a total of four (two male and two female) adult volunteers with varying hand surface areas (Table S1), to reflect incidental soil ingestion pathways of children. This study was reviewed and approved by the Taipei Medical University-Joint Institutional Review Board (approval no: 201101026). All four volunteers signed a consent form. The hand-pressing trials were conducted to determine amounts of soil transferred to the hand. In contrast, the hand-to-mouth transfer ratio was expressed as the mass fraction of the total hand load transferred to the mouth by three types of mouthing activities. Key factors that could influence hand soil loading and the hand-to-mouth transfer ratio were examined as follows.

1. 1) Soil texture: Soils with two textures were selected, including sand (i.e., 0.05–2.0 mm) and clay (i.e., ≤0.002 mm). In addition, standard soils (Nacalai Tesque, Inc., Kyoto, Japan) were commercially obtained and tested.

2. 2) Contact pressure: Three contact pressures of 0.1, 0.5, and 1.0 psi, were tested, following Hubal et al. (2008).

3. 3) Contact time: The contact time for the soil-to-hand press tests was set to 3 s based on Beamer et al. (2008), and the hand-to-mouth contact time was set to 1 s based on our previous study (Tsou et al., 2017).

4. 4) Standard sand and clay was selected to examine the dependence of soil loading and hand-to-mouth transfer on the moisture content of the soil. The moisture content was 0.1% of the original standard sand. On the basis of Das (2009), which showed that the liquid limit (LL) of a typical clay sample was 50.0% and the plastic limit (PL) was 30.9%, four moisture contents were applied to the soil including 10% (i.e., the moisture content of the original standard clay), 30%, 50%, and 60%. The moisture content was calculated by the following equation:

$$\theta_w = W_w / W_s \quad (1)$$

where θ_w is the unitless weight ratio of water to soil, W_w is the water weight, and W_s is soil weight.

2.2 Hand-pressing trails

Hand soil loadings from hand-pressing trials were determined based on methods described by Kissel et al. (1998a). The main steps of this protocol included:

1. 1) Washing a subject's hands thoroughly, then air-drying with a blower for 20 s.

2. 2) Hand pressing (with the palm down and fingers spread) onto a shallow pan containing autoclave-sterilized soil for 3 s with pressing pressures of 0.1, 0.5, and 1.0 psi.

3. 3) Washing the soils from both hands and collecting the wash water in a plastic bottle.

4. 4) Performing each of the three contact pressures by each of the four adult volunteers in triplicate.

2.3. Hand-to-mouth transfer

The highest frequency of children's hand-to-mouth activities are from thumb-sucking, finger-sucking, and palm-licking (Kissel et al., 1998a); therefore, similar activities were performed by study subjects to examine hand-to-mouth soil transfer. When the hand-to-mouth experiments were performed, four soil aliquots from thumb-sucking, finger-sucking, palm-licking, and the remaining soil on the hand were collected. Descriptions of the experimental protocols are given below.

1. 1) A subject's right hand was thoroughly washed and then air-dried with a blower for 20 s.

2. 2) The hand was pressed (with the palm down and fingers spread) onto a shallow pan of autoclave-sterilized soil for 3 s with downward pressures of 0.1, 0.5, and 1.0 psi.

3. 3) The index, middle, and ring fingers above the first knuckle were put into the mouth, and the mouth was rinsed three times to collect the first soil aliquot.

4. 4) Then, the thumb was sucked, and the mouth was rinsed three times to collect the second soil aliquot.

5. 5) Then, the palm was licked, and the mouth was rinsed three times to collect the third soil aliquot.

6. 6) The remainder of the soil was washed from the hand into a plastic bottle to collect the fourth soil aliquot.

7. 7) Performing each of the three contact pressures by each of the four adult volunteers in triplicate.

2.4. Measurement of soil loadings

Soil loadings resulting from soil-to-hand pressing and hand-to-mouth transfer activities were determined by an established weighing analytical procedure. To obtain the total mass of soil in the collected aliquots, standard method NIEA-W210.57A from The Environmental Analysis Laboratory of Taiwan EPA was performed (TEPA, 2013). Since the solid weight of the hand washing samples could possibly be less than the method detection limit (i.e., 2.5 mg), liquid samples were first centrifuged and concentrated, followed by filtering and drying, and then the solids collected were weighed on an ultra-microbalance. The detailed analytical procedures were as follows:

1. 1) Hand-washing samples were concentrated from a plastic bottle and transferred to a 15-mL centrifuge tube.

2. 2) A concentrated soil sample was centrifuged at 3500 rpm for 30 min.

3. 3) Soil from the bottom of the tube was transferred to an aluminum (Al) evaporating pan.

4. 4) Soil in the Al evaporating pan was dried in a 105 °C oven for >24 h.

5. 5) The final sample was weighed using an ultra-microbalance.

2.5. Soil loading determination

After the soil amount was determined, the soil loading was calculated based on the following equation:

$$AF_i = SL_i/SA \quad (2)$$

where AF_i is the hand soil loading (mg/cm^2), SL_i is the amount of soil washed from the hand (mg), and SA is the surface area of the hand (cm^2).

The hand surface area of adult volunteers was calculated by two methods. Traced hand surface areas of adult volunteers were calculated by a method proposed by Leckie et al. (2000); the calculation is shown in Eqs. (3), (4), (5), (6), (7). A volunteer pressed a hand onto a hand tracing template, and the surface area was estimated using AutoCAD LT® 2014 (Autodesk, San Rafael, CA, USA). The traced hand surface area (THSA) was estimated by assuming that one finger surface area (FSA) can be represented by a cylinder's surface area (i.e., the finger width (FW) was the diameter and finger length (FL) was the height) plus a round surface area (of the fingertip). The FSA was calculated from the following equation:

$$FSA_1 = \left(\pi \cdot \quad FW_i \cdot FL_i\right) + \left(\frac{\pi FW_i^2}{4}\right) \quad (3)$$

where FSA_i is the surface area of each finger (cm^2), FW_i is the widest point of each finger (cm), and FL_i is the length of each finger (cm).

The sum of the surface area from five fingers refers to the total finger surface area (TFSA), calculated by the following equation:

$$TFSA = \sum_{I=1}^{5} FSA_1 \quad (4)$$

Leckie et al. (2000) also assumed that the palm is a cylinder (not including the finger surface area); the palm area was also estimated with AutoCAD LT 2014 (Autodesk). The palm area is the same as the back area of the hand, the width of the middle finger is the height, the thickness of the palm is the same as the width of the middle finger, and the side of the palm without connecting fingers and wrist is the exposed area. The palm exposure perimeter (PEP) was calculated by the following equation:

$$PEP = TPP - \left(\sum_{I=1}^{5} FW_i + WW\right) \quad (5)$$

where PEP is in cm, TPP is the total palm perimeter (cm), and WW is the wrist width (cm).

The total palm surface area (TPSA) was calculated by the following equation:

$$TPSA \ = \ (2 \times PSA) + MFW \times PEP \quad (6)$$

where TPSA is total palm surface area (cm^2), PSA is the palm surface area (cm), and MFW is the middle finger width (cm).

Finally, the THSA is the sum of the TFSA and TPSA, calculated as:

$$THAS = TFSA + TPSA \quad (7)$$

The entire total hand surface area (i.e., both hands) of adult volunteers was calculated by Eqs (8), (9) for males and females, respectively, as proposed by Anderson et al. (1985):

$$WHSA_{male} = 0.0257 \quad \cdot W^{0.573} \cdot H^{-0.218} \quad (8)$$

$$WHSA_{male} = 0.0131 \quad \cdot W^{0.412} \cdot H^{-0.0274} \quad (9)$$

where $WHSA_{male}$ is the whole hand surface area for males (m^2), W is the body weight (kg), H is the height (cm), and $WHSA_{female}$ is the whole hand surface area for females (cm^2).

The weight, height, and hand surface areas estimated by the two methods for the four adult volunteers are summarized in Supplementary Table S1.

2.6. Soil particle size analysis

After determining soil loadings, a concentrated sample was put into a 50-mL centrifuge tube with water; a dispersant (i.e., sodium hexametaphosphate) was then added, and the resulting samples were screened with a 35-mesh (500 μm) standard sieve. The sample was then analyzed by a particle size analyzer (Cilas 1090) to investigate variations in the particle size distribution, expressed as a number frequency, when the soil was transferred from the soil to the hand and from the hand to the mouth. Particle diameters in volume were subsequently determined based on the size distribution results.

2.7. Statistical analysis

Descriptive statistical methods were used to describe soil loadings in the hand-pressing trials. Because the distributions of the soil-loading data and transfer ratios were right-skewed, non-parametric statistical tests were used. The Mann-Whitney *U* test was used to test significant differences of binomial variables. The Kruskal-Wallis test was used to assess significant differences in trinomial variables. Multiple linear regression models were run to determine relationships between hand-pressed soil loadings by soil type, pressing pressure, and moisture content. Statistical analysis was carried out using SPSS (vers. 20 for Mac; SPSS, Chicago, IL, USA). Results were considered significant in a two-sided test if $p < 0.05$.

3. Results and discussion

3.1. Factors affecting soil loadings in hand-pressing trials

Geometric means and geometric standard deviations (SDs) of hand-pressed soil loadings are presented in Table 1. For soils with different textures, a greater amount of sand adhered than

clay (original soil) at all three contact pressures. For example, at a contact pressure of 1.0 psi, the geometric mean (geometric SD) of measured sand loading, based on the traced hand area method, was 0.57 (1.84) mg/cm^2, compared to 0.33 (1.39) mg/cm^2 for clay (original soil). Ferguson et al., 2009a, Ferguson et al., 2009b conducted tests in a computer-controlled chamber and showed that the amount of adhered sand was significantly greater than for lawn soil; contact pressure levels were 0.2, 0.3, 0.4, and 0.5 psi and contact times were 10, 30, and 50 s. Although the experimental test parameters and measurement techniques between our and Ferguson's studies differed, the same trend of the dependence of soil loadings on the pressing pressure was still observed.

The dependence of soil loadings on moisture contents of clay is also shown in Table 1. The geometric mean (geometric SD) soil loadings of clay based on the traced hand area method at moisture levels of 10% (i.e., original soil), 30%, 50%, and 60% at 1.0 psi were 0.33 (1.39), 0.004 (3.08), 0.49 (1.66), and 16.4 (1.20) mg/cm^2, respectively. This U-shaped relationship (Fig. 1) between soil loadings of clay and moisture content, with 60% and 30% moisture contents having the largest and smallest soil loadings, respectively, was mainly due to the fact that the moisture of the clay was approaching its liquid and plastic limits. A moisture content of 60% for clay exceeds the liquid limit, which is at approximately 50%, causing the tested clay to be in a liquid state like mud (Brady and Weil, 2008), thus leading to much greater adherence to the hands following performance of hand-pressing. On the contrary, the plastic limit of the clay that we tested was at a 30.9% moisture content. The clay in this plastic state at a 30% moisture content had the strongest soil cohesion, thus revealing the smallest amount of soil loadings on the hands after pressing on it.

Table 1 also shows soil loadings estimated based on the traced hand area method. In general, these results were greater than those estimated based on the whole hand area method. This is assumed to have been due to larger hand area estimates based on the method provided in Anderson et al.'s (1985) study (Table S1). Additionally, soil loadings in Kissel et al. (1998a) were 0.04–0.35 mg/cm^2 (calculated by Eqs. (3), (4)), which were smaller than those we found during the sand trials. Differences in soil loadings between the two studies could have been due to differences in textures of the soils used in these studies. In the current study, we used commercially obtained standard sand, but Kissel et al. (1998a) used a locally obtained natural loamy sand.

Significant differences were not observed in soil loadings for sand ($p = 0.273$) and clay (original soil) ($p = 0.294$) at the tested pressures of 0.1, 0.5, and 1.0 psi (Table 1). However, soil loadings had positive correlations with contact pressures at all moisture contents, except for the original clay. These results suggest that a larger contact pressure led to greater soil adherence for clay soil at elevated moisture contents. Significant differences were also found in soil loadings between different contact pressures ($p < 0.05$) (Table 1).

To evaluate the effects of soil type (sand and clay (original soil)) and pressing pressure (0.1, 0.5, and 1.0 psi) on soil loadings, multiple regression model 1 was conducted (Table 2). Results of the regression model for the object-to-mouth contact frequency are shown in Table 2. The results indicated that the soil type significantly affected the soil loadings on the hands ($p < 0.001$) after controlling for hand-pressed pressures. That is, the type of soil was

more important in influencing soil loadings on the hands than was the pressing pressure. We then evaluated the effects of both the moisture content of the clay soil tested (i.e., original clay soil (10%), 50%, and 60%) and the hand-pressing pressures on the soil loadings. Results of model 2 indicated that the moisture content of clay ($p < 0.001$) and pressing pressure ($p = 0.004$) were both significantly associated with the measured loadings of clay soil (Table 2). We also examined the correlation between soil loading and moisture as well as with the pressing pressure. We found that soil loadings were better correlated with moisture levels ($r = 0.885$, $p < 0.001$) than hand-pressing pressures ($r = 0.084$, $p = 0.183$) (Table 2).

3.2. Soil particle size variations after soil-to-hand transfer

Table 3 shows a summary of particle size distributions for soils after the hand-pressing trials. Results from Table 1, Table 3 suggest that all soil loadings observed, except for that from the 60% moisture clay results, were well below the estimated monolayer coverage. The median particle sizes of sand at contact pressures of 0.1, 0.5, and 1.0 psi were determined to be 121, 120, and 127 μm, respectively; these were quite similar to the particle size of the original sand (i.e., 124 μm) (Table 3). Fig. S1 also displays particle size distributions of the original sand and those of sand that adhered to the hand after hand-pressing trails at three pressures. The experimental results indicated that the difference between the particle size distributions of the original sand and those of sand that adhered to the hand after hand-press trials was insignificant.

The particle size change between original clay and that transferred from hand pressing as a function of the change in pressure showed different results than those found from identical experimental procedures using sand (Table 3, Fig. S2). Median particle sizes of transferred clay at 0.1, 0.5, and 1.0 psi were 10.0, 9.2, and 9.0 μm, respectively, which were much larger than that found for the original clay (i.e., 5.8 μm). A significant peak shift in the size distribution of transferred clay particles after the hand-pressing trails was clearly observed based on results obtained by the particle size analyzer. These data suggest that hand-pressing and the pressure exerted may have enhanced adherence of clay particles of larger sizes to the hand. It is important to note that the increase in the median sizes of the transferred soil particles might not have stemmed from the aggregation of clay particles by hand-pressing or the pressure exerted, because a dispersant was employed during the particle size examination by the laser particle analyzer. Also, the dependence of the resulting median and percentile particle sizes on the pressing pressure was less pronounced.

To further understand the influence of the moisture content on variations in the particle size distribution after soil transfer, clay with different moisture contents was evaluated in the hand-pressing trial at 1.0 psi (Table 3, Fig. S3). The median size of transferred soil particles with a 60% moisture content at 1.0 psi was 16.1 μm. This was larger than the median size of original clay particles at 9.0 μm transferred at 1.0 psi. The median particle size of soil transferred by hand-pressing being larger than that of the original clay could also have been due to enhanced adherence of larger clay particles to the hand due to hand-pressing.

We found that half of the sand particles which adhered to the hands were smaller than 127 μm, and 90% of total sand particles that adhered to the hands was smaller than about 200

μm. We also found that large clay particles preferably adhered to the hands when the moisture of clay soil was high at 60%. Data summarized in Table 4 also suggest that the sizes of most particles that adhered to the hand were smaller than 150 μm; thus, smaller-sized particles tended to remain on the skin surface of the hands. Kissel et al. (1996a) and Choate et al., 2006a, Choate et al., 2006b also found that soils with a higher moisture content had large particle fractions adhering to the hands compared to those with low and medium moisture contents (Table 4). Our experimental results are close to those from previously published studies.

3.3. Hand-to-mouth soil-transfer ratios

Fig. 2 illustrates the mean hand-to-mouth transfer ratios of sand and clay particles following three types of mouthing activities. Distributions for hand-to-mouth transfer ratios are shown in Supplementary Table S2. Among the three mouthing activities, palm-licking had the highest transfer ratios for sand particles, namely 16.2%~17.2%, at all pressing pressures. In contrast, the highest transfer ratios at all pressing pressures for clay particles were from finger-mouthing (i.e., 10.4%~16.1%). Thumb-sucking resulted in lower hand-to-mouth transfer ratios for sand (9.14%~10.4%) and clay (5.48%~10.7%) particles. Significant differences were observed for clay particle transfer ratios between different pressing pressures for finger-mouthing ($p < 0.05$) and palm-licking ($p = 0.001$).

Fig. S4 shows transfer ratios for clay particles obtained from three types of mouthing activities at different moisture contents. The finger-mouthing activity resulted in the highest transfer ratio, while thumb-sucking led to the lowest transfer ratio, under all of the moisture contents tested. Significant differences were only observed between transfer ratios under various mouthing activities only for original clay ($p < 0.05$).

The experimental results suggest that the pressing pressure and moisture content of the soil may have negative correlations with soil-transfer ratios. Because a higher pressing pressure and higher moisture content appeared to cause larger soil loadings on the hand, transfer ratios then tended to decrease. To our knowledge, only Kissel et al. (1998a) conducted related research on skin-to-hand transfer and obtained hand-to-mouth transfer ratios. Table 5 compares our results with those reported in Kissel et al. (1998a). Results from these two studies are similar, indicating that finger-mouthing and palm-licking caused larger transfer ratios than thumb-sucking. However, transfer ratios obtained in the present study were smaller than those shown in Kissel et al. (1998a). The differences could have been due to different soil textures and soil loadings between the two studies. The soil loadings in Kissel et al. (1998a) were 0.04–0.35 mg/cm^2, predominantly smaller than those obtained in our study (Table 1). Additionally, transfer ratios from palm-licking of clay particles were smaller than those for finger-mouthing, which may have been because finer particles more readily adhered to the skin of the palm and were transferred from the hand to the mouth with greater difficulty; the surface of the palm has more wrinkles than do the thumb and fingers.

3.4. Variations in soil particle sizes after hand-to-mouth transfer

The median and percentile particle sizes of soil particles transferred from the hand to the mouth are summarized in Tables S3 and S4. The experimental results showed that particle

sizes of transferred sand after the three mouthing activities (i.e., palm-licking, finger-sucking, and thumb-sucking) were smaller than those for the original sand and hand-pressed transferred sand. The particle size distribution shown in Fig. 3a also indicated generation of a second mode at sizes of 10–100 μm. However, for clay, particle sizes of the transferred clay after the three mouthing activities were all larger than those for the original and hand-pressed clay (Fig. 3b). Median particle sizes of sand and clay particles transferred after hand-to-mouth activities were 71–106 and 16.1–28.5 μm, respectively. Differences in particle sizes of soils among the three mouthing activities were insignificant.

Although the particle size of transferred clay from the hand to the mouth was larger than those of the original and hand-press clays, the median particle size was still smaller than 50 μm. Some studies discovered that heavy metals at higher concentrations were present in finer particle size fractions (i.e., sized <50 μm), and that the concentration of metals in soils increased with a decreasing particle size (Ljung et al., 2006; Wang et al., 2006). Although with the exception that Bergstrom et al. (2011) showed that finer soil particles may contain contaminants, such as Cd and Zn at lower concentrations in smaller soil particles, the shift to smaller particle sizes of soil adhered to the hand may enhance the harmful effects on children's health. A better understanding of the magnitude, speciation, and bioavailability of toxic substances in soil particles of different sizes is undoubtedly critical to improve the risk assessment results for children engaged in mouthing behaviors.

It is also critical to address that hand-pressing trial results from different studies showed large variations. For example, Bergstrom et al. (2011) conducted a soil-handling protocol with adult volunteers handling 1.5 cups of each unfractionated medium sample in a plastic bag for 30 s under wet and dry conditions. The results suggested that they may have underestimated potential exposures to soil contaminants. This may have been because the protocol might not have reflected actual concentrations adhering to the hands when contacting whole, unfractionated medium samples. Other field trials also revealed lower amounts of soil loadings during subsequent activities, abrasion, and movements compared to the initial contact (Kissel et al., 1998b; Shoaf et al., 2005). In contrast, the hand-pressing trails in our study may have overestimated exposures to soils, but this overestimation may be considered a worst case for assessing exposures and health risks of children living near contaminated sites to ensure that children are well protected.

There are several limitations of our study. For this control laboratory-scale experiment, four adult volunteers were used as surrogates for children under 6 years old in order to avoid the risk of children accidentally swallowing the test soils. Thus, the results might not well represent actual soil loadings or soil transfer rates for children. Our sample size of volunteers and soil types and conditions were limited. Additional research on this topic both in a lab setting and as part of observational studies involving children is recommended.

4. Conclusions

Soil loadings under various test conditions were successfully obtained through laboratory-scale hand-pressing investigations. Factors influencing soil loadings and the change in

particle sizes after soil transfer were also examined. Key findings are summarized as follows.

1. 1) The texture of the soil influences its adherence to skin; sand exhibited higher soil loadings than clay.
2. 2) The moisture content of the soil greatly influences adherence values; when clay contained moisture exceeding its liquid limit, soil loadings to the skin significantly increased.
3. 3) Greater pressure led to higher soil adherence values.
4. 4) Sizes of particles transferred from the soil to the hand and the hand to the mouth were mainly smaller than 150 μm.
5. 5) Mouthing behaviors can lead to an increase in clay particle sizes transferred to the mouth.

Soil loadings and soil transfer ratios are important parameters influencing both the exposure and dose from contacting and ingesting soil particles. The data provided here are of great importance for future health risk assessments, especially for children who exhibit significant mouthing behaviors. A better understanding of variations in soil particle sizes after transfer from soil-to-hand or hand-to-mouth activities can avoid underestimating exposure risks when bulk concentrations of contaminants in whole soil are used instead during health risk assessments.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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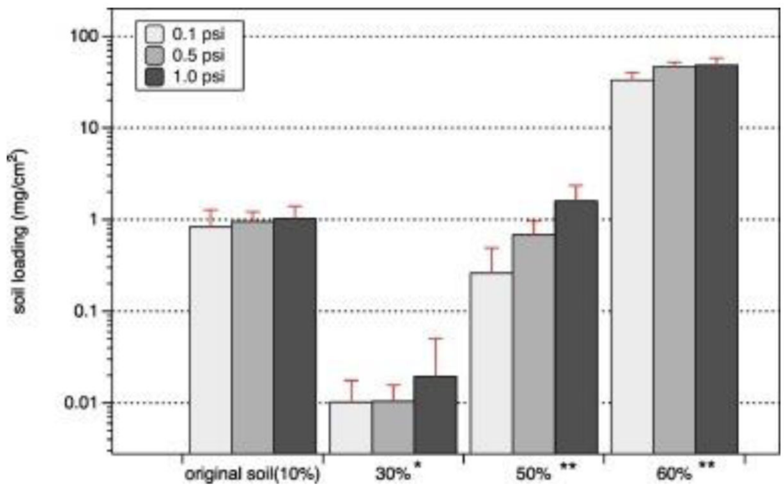
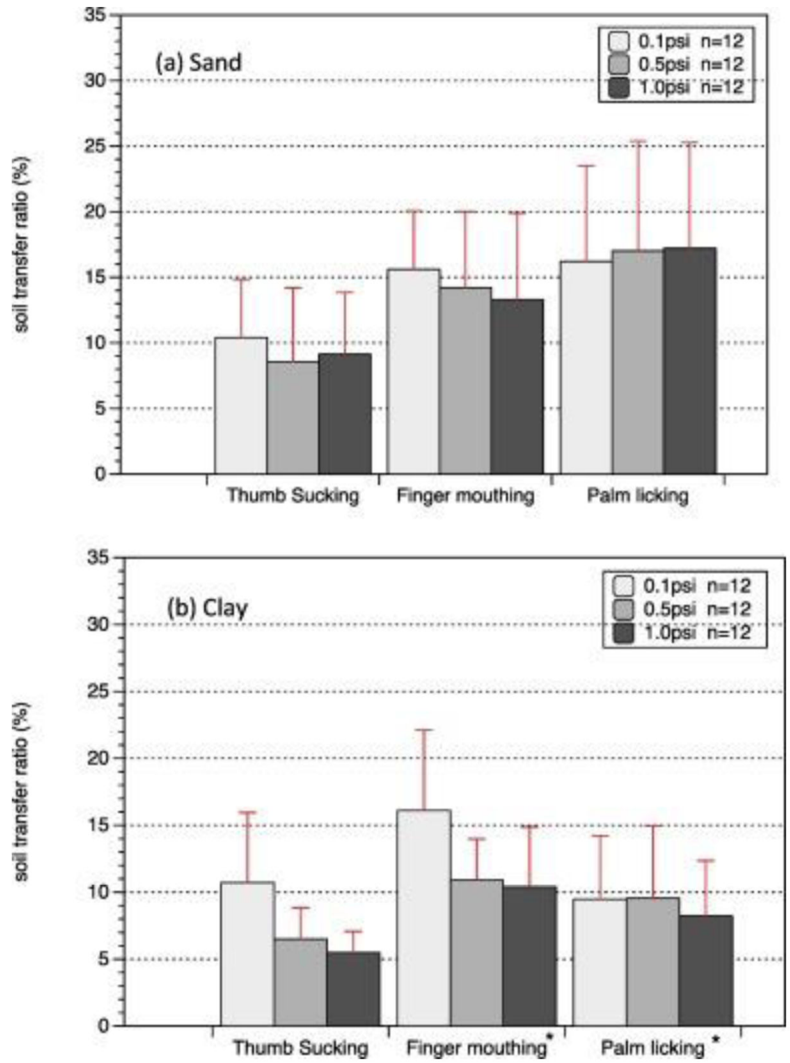


. Soil loadings for clay at different moisture contents in the hand-pressing trial. * $p < 0.05$, ** $p < 0.001$.



. Hand-to-mouth transfer ratios of (a) sand and (b) clay particles following three types of mouthing activities. * $p < 0.05$.

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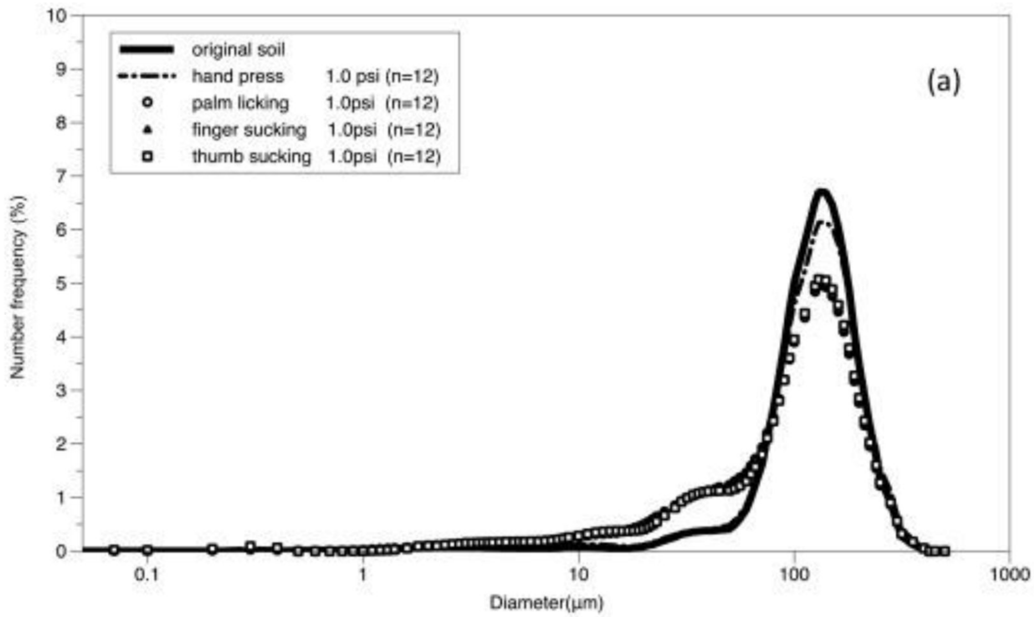

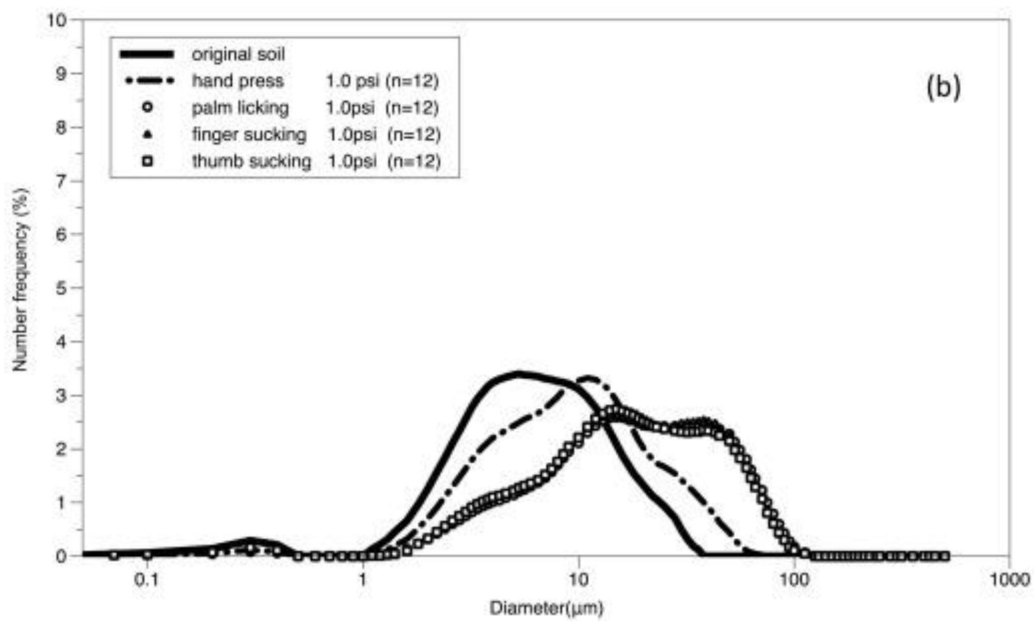

. Particle size distributions of (a) sand and (b) clay particles prior to and after mouthing activities at 1.0 psi.

Table 1.

Geometric mean (geometric standard deviation) of calculated soil loadings based on hand-pressing trials.

Study	Soil texture	*n*	Soil loading based on traced hand surface area[a] (mg/cm^2)				Soil loading based on whole-hand surface area[b] (mg/cm^2)			
			0.1 psi	**0.5 psi**	**1.0 psi**	***p*value[c]**	**0.1 psi**	**0.5 psi**	**1.0 psi**	***p*value**
This study	Sand	12	0.45 (1.89)	0.68 (2.10)	0.57 (1.84)	0.273	0.41 (1.88)	0.63 (2.06)	0.52 (1.81)	0.296
	Clay[d](original soil)	12	0.25 (1.69)	0.32 (1.30)	0.33 (1.39)	0.294	0.23 (1.68)	0.29 (1.30)	0.31 (1.67)	0.387
	Clay (30% moisture)	40	0.003 (2.59)	0.003 (2.06)	0.004 (3.08)	0.026	0.002 (2.68)	0.003 (2.13)	0.004 (3.11)	0.039
	Clay (50% moisture)	12	0.069 (2.02)	0.22 (1.52)	0.49 (1.66)	<0.001	0.064 (2.07)	0.201 (1.52)	0.45 (1.62)	<0.001
	Clay (60% moisture)	20	11.2 (1.24)	16.0 (1.12)	16.4 (1.20)	<0.001	10.3 (1.26)	14.7 (1.13)	15.1 (1.18)	<0.001
Kissel et al. (1998a)	Loamy sand	36					0.04–0.35[e]			

[a] Soil loading based on the traced hand surface area was estimated based on the palmar hand area according to the method of Leckie et al. (2000).

[b] Soil loading based on the whole-hand surface area was estimated based on the total hand area using the formula provided in Anderson et al. (1985).

[c] Kruskal-Wallis test, significance level: $p < 0.05$.

[d] The moisture content of original clay was approximately 10%.

[e] Average moisture contents ranged 0.8%~1.6%. Soils were sieved to 2 mm. There was no description of pressing pressures used in Kissel et al. (1998a). Data are presented in terms of a range of measured soil loadings.

Table 2.

Multiple regression model of hand-pressed soil loadings on three explanatory variables: soil type, hand-pressed pressures, and soil moisture.

Factor	Model 1[a]			Model 2[b]		
	Beta	*p* value	Adjusted R^2	Beta	*p* value	Adjusted R^2
Intercept	1.001	<0.001	0.568[f]	−9.322	<0.001	0.790[f]
Soil type[c]	−0.347	<0.001				
Pressure[d]	0.020	0.339		0.762	0.004	
Moisture[e]				6.936	<0.001	

[a] Model 1 included soil types and pressures for predicting soil loadings for sand and clay (original soil).

[b] Model 2 included pressures and moistures for predicting soil loadings for clay.

[c] Two soil groups: 1: sand; 2: clay (original soil).

[d] Three pressure groups: 1: 0.1 psi; 2: 0.5 psi; 3: 1.0 psi.

[e] Three clay soil moisture groups: 1: original clay (10%); 2: 50%; 3: 60%.

[f] $p < 0.01$.

Table 3.

Median, 10th (P10), and 90th (P90) percentile soil particle sizes after hand-pressing trials for sand and clay (with moisture contents of 10% and 60%).

Pressure (psi)	*n*	Particle size (μm)		
		Median	**P10**	**P90**
Sand (original)[a]		124	64	199
0.1 psi	12	121	45	204
0.5 psi	12	120	46	202
1.0 psi	12	127	57	209
Clay (original)[a,b]		5.8	2.0	15.9
0.1 psi	12	10.0	2.8	30.6
0.5 psi	12	9.2	2.8	27.9
1.0 psi	12	9.0	2.8	26.4
Clay (60%)				
0.1 psi	12	15.9	3.4	44.8
0.5 psi	12	16.2	3.4	45.7
1.0 psi	12	16.1	3.4	45.4

[a] The median and percentile particle sizes are for the original soil prior to hand-pressing trials.

[b] The moisture content of the original clay was approximately 10%.

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Table 4.

Summary of available results from studies on adhered soil particle size variations after hand-pressing trials.

Study	Study type and conditions	Particle size (μm)	
		Median	P90[b]
This study[a]	Laboratory: sand	127	209
	Laboratory: clay	9	26
Duggan et al. (1985)	Laboratory	57	130
Kissel et al. (1996a)	Laboratory: dry soil	62	210
	Laboratory: wet soil	150	350
Choate et al. (2006b)	Laboratory: low-moisture soil	33	110
	Laboratory: medium-moisture soil	44	120
	Laboratory: high-moisture soil	80	220
Yamamoto et al. (2006)	Laboratory: garden soils	–	–
	Field: children	67	134
Siciliano et al. (2009)	Laboratory: agricultural soils	40	370
	Laboratory: brownfield soils	125	760
	Field: Iqaluit residents	40	130
Ikegami et al. (2014)	Field: children	38[b]	–

[a] Particle sizes of sand and clay particles transferred to the hand were obtained at a contact pressure of 1.0 psi.

[b] P90, 90th percentile.

Table 5.

Comparison of the ratio of measured soil transfer results to values reported by Kissel et al. (1998a).

Study	Soil texture (moisture content)	Soil transfer ratio (%)		
		Geometric mean (geometric standard deviation)		
		Thumb-sucking	Finger-mouthing	Palm-licking
This study[a]	Sand	7.71 (2.22)	10.90 (2.22)	15.00 (1.59)
	Clay (original, 10%)	5.26 (1.48)	9.61 (1.61)	7.33 (1.73)
	Clay (50%)	1.37 (2.19)	2.74 (2.71)	2.35 (1.52)
	Clay (60%)	2.43 (1.60)	4.05 (2.56)	3.72 (1.80)
Kissel et al. (1998a)	Loamy sand (0.8%~1.6%)	10.10	15.90	21.90

[a] Contact pressure: 1.0 psi.